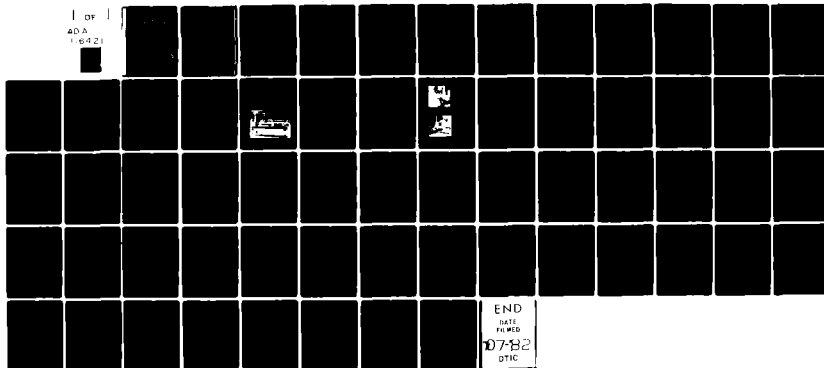AD-A116 421   SOUTHWEST RESEARCH INST SAN ANTONIO TX ARMY FUELS AN--ETC F/G 21/2
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EFFECT OF FLAME TEMPERATURE AND FUEL COMPOSITION ON SOOTING TENDENCY IN A RESEARCH COMBUSTOR

INTERIM REPORT
AFLRL No. 158

By

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December 1981

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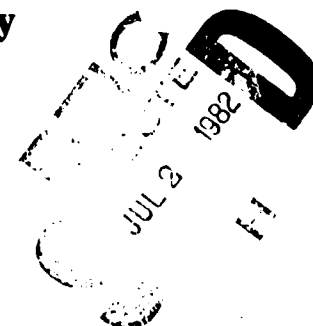| REPORT DOCUMENTATION PAGE		READ INSTRUCTIONS BEFORE COMPLETING FORM
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Turbine Combustor	H/C Ratio	Fuel Sensitivity
Fuel Properties	Flame Temperature	

20. ABSTRACT *(Continue on reverse side if necessary and identify by block number)*

Several fuel blends containing alkyl benzenes, methyl naphthalenes, tetralin and Indene were prepared with hydrogen contents ranging from 11.5 to 14.2 percent. The effects of burner inlet conditions on the sooting tendency of the test fuels were measured in a Phillips 2-inch diameter cylindrical combustor capable of inlet pressures and temperatures up to 1.6 MPa and 1030K, respectively. Both flame radiation and opacity measurements were used to determine the soot formed in the primary zone of the burner. Flame

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20. ABSTRACT (Cont'd)

temperature was measured by the Kurlbaum method. Combustion efficiency and fuel/air ratio were determined from gaseous emissions.

A sensitivity of sooting tendency to H/C ratio was determined from the correlation of flame radiation intensity with H/C ratio. This sensitivity varied significantly with operating parameters such as burner inlet temperature and reference velocity. The effects of polycyclic aromatics were determined by comparing the sensitivity to H/C ratio of fuels containing alkyl naphthalenes and tetralin to that of the fuels blended with alkyl benzenes.

The increased sooting tendency of fuels containing alkyl naphthalenes and tetralin was most strongly dependent on the fuel/air ratio and the reference velocity. Soot concentrations in the primary zone of the burner increased as the flame temperature was raised. Flame temperature was found to be important in sooting tendency, but it did not explain the effects of H/C ratio. The H/C ratio appears to influence the flame chemistry, especially the role of OH radicals in oxidizing soot precursors.

SUMMARY

Soot formation from a variety of jet fuels containing alkyl benzenes, alkyl naphthalenes, tetralin, and indene, with hydrogen contents ranging from 11.5 to 14.2 percent has been studied over a wide range of combustor conditions covering the operating envelopes of many gas turbine engines. These results may explain why engines show different sensitivities to H/C ratio of the fuel and why a few are sensitive to the molecular structure, e.g., poly-cyclic aromatics, while most are not.

The effects of burner inlet conditions on the sooting tendency of the test fuels were measured in a Phillips 2-inch diameter cylindrical combustor capable of inlet temperatures and pressures up to 1030K (1395°F) and 1.6 MPa (232 psia), respectively. Both flame radiation and opacity measurements were used to determine the soot formed in the primary zone of the burner. Flame temperature was determined as a soot reversal temperature using the Kurlbaum method. Combustion efficiency and fuel/air ratio were determined from gaseous emissions.

A sensitivity of sooting tendency to H/C ratio was determined from a correlation of flame radiation with H/C ratio. Three sensitivities to H/C ratio were determined: S_1, based on fuels containing alkyl benzenes; S_2, based on fuels containing tetralin; and S_3, based on fuels containing alkyl naphthalenes.

The sensitivity, S_2, to H/C ratio for fuels blended with tetralin was the same as S_3 which used fuels blended with adding methyl naphthalenes. Note that, to lower the H/C ratio a given amount, tetralin must be added in larger quantities than methyl naphthalene. The S_1 sensitivity for fuels containing alkyl benzenes was less at most operating conditions.

The S_1 sensitivity varied with operating conditions as follows:

- Decreased rapidly as burner inlet temperature increased.
- Increased rapidly as reference velocity increased.

- Changed gradually with fuel/air ratio.
- Was only weakly dependent on inlet gas density.

Qualitatively, the same trends were observed for fuels containing large amounts of alkyl naphthalenes and tetralin. The difference in sensitivities, i.e., the increased sooting due to the molecular structure effect, was equivalent to lowering the hydrogen content as much as 1 percent, depending mostly on fuel/air ratio and reference velocity.

The increase in soot formation caused by polycyclic aromatics was directly proportional to the polycyclic aromatic ring carbon in the fuel.

The temperature dependence (E $\approx$ 50 kcal/mole) of soot formation obtained was of the same magnitude as that obtained in other independent studies on the formation of soot from acetylene pyrolysis.

When the hydrogen content of the fuel was reduced from 14.2 to 11.5 percent, the soot increased by a factor of 2.5, while the temperature increased only about 35K. An attempt to explain the increased sooting by temperature effects alone would require an effective activation energy of 275 kcal/mole for these experimental conditions, much greater than the 50 kcal/mole measured for the temperature effect alone. Chemical effects other than temperature appear to be responsible for the dependence of sooting tendency on hydrogen content.

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FOREWORD

ACKNOWLEDGMENTS

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I. INTRODUCTION

Gaining a more basic understanding of soot formation in gas turbine com-
bustors has become increasingly important because future fuels may vary
significantly in composition, and specifications may need to be changed to
ensure an adequate fuel supply. Aside from the environmental aspects, and
possible signature problems on military aircraft, the greatest concern about
soot in gas turbines is the effect it has on combustor liner durability.
Incandescent soot particles transfer radiant energy from the flame to the
combustor liner. The air required to cool the liner is largely determined
by this radiative heat flux; combustor design is critical because too much
cooling air may quench combustion intermediates and adversely affect low
power emissions. Several studies (1-5)* have shown that both the flame
radiation intensity and increases in the combustor liner temperature corre-
late consistently with decreased fuel hydrogen content. For a typical
engine such as the J79, increased liner temperature due to a reduction in
fuel hydrogen content from 14.5 to 12 percent will reduce the cycle life by
65 percent. (6) The correlation of soot formation with hydrogen content, or
H/C atom ratio, has recently been extended to include not only petroleum-
based fuels, but also fuels derived from oil shale, coal, tar sands, emul-
sions containing both water and alcohols, and solutions of alcohols and jet
fuels. (4, 5, 7) Based on these studies, it has been concluded that H/C
ratio more consistently predicts sooting tendency than aromatic content,
aromatic ring carbon content, or smoke point.

The effects of physical properties on soot formation have also been inves-
tigated. Increases in viscosity and boiling-point distribution increase
droplet lifetime which, in some experiments, has been shown to increase
sooting. (8) However, experiments in this laboratory (4) with a Phillips
2-inch high-pressure research combustor and a T63 combustor have shown that
significant variations in viscosity and boiling point distribution do not
increase the tendency of fuels to soot any more than would be expected from
their hydrogen/carbon ratio. This precludes the possibility of liquid-phase
pyrolysis and supports a gas-phase soot formation process.

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*Underscored numbers in parentheses refer to the list of references at the
end of this report.

Recently, Glassman and Yaccorino (9) have shown the importance of flame temperature on soot formation in laminar diffusion flames. They found that soot is formed more readily at higher flame temperatures because the pyrolysis reactions which make soot precursors have a greater temperature dependence than the oxidation reactions that destroy virgin soot material. Millikan (10) found the temperature dependence of soot formation in premixed flames to be opposite of that in diffusion flames. This is believed to be due to the strong temperature dependence of the OH radical concentration in premixed flames which appears to play a key role in the oxidation of soot precursors.

In view of the rather marked effect of flame temperature on soot production in laminar diffusion flames, it has become suspect that the H/C ratio soot correlation is simply an artifact caused by variations in flame temperature with fuel composition. Note that the adiabatic flame temperature increases as H/C ratio is reduced; this is in accordance with the dependence of soot production on flame temperature in diffusion flames.

However, if the only effect of changing H/C ratio was to change the flame temperature, the effect of H/C ratio on soot concentration in the primary zone of a combustor should not vary significantly with operating parameters such as pressure and reference velocity, which have a relatively small effect on flame temperature. On the contrary, work in this laboratory (4) and the work by Friswell (3) indicates that the sooting tendency in gas turbine combustors can be much more sensitive to H/C ratio at one operating condition than another. Similarly, the effects of H/C ratio on sooting tendency are difficult to explain for engines of different design. While the sooting tendencies of most gas turbine engines correlate well with H/C ratio and also show a similar dependence, some engines exhibit a much greater sensitivity to H/C ratio than others.(11) Figure 1 shows that the smoke tendency of the J85 engine is particularly more sensitive to H/C ratio than other well-known designs.

These effects of operating parameters and combustion design on the sensitivity of sooting tendency to H/C ratio do not seem to be consistent with the theory that H/C ratio only affects the flame temperature. Instead,

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it seems more probable that H/C ratio plays a key role in some major aspect of soot chemistry.

While H/C ratio has been found to be the most important indicator of the sooting tendency, the molecular structure is sometimes a significant factor as well. To measure the sooting tendency of a test fuel containing some unusual component, the test fuel is compared with a typical petroleum-base fuel of equal hydrogen content. The typical petroleum base fuel is, for the most part, a blend of paraffins and single ring aromatics. In a recent study (12), six test

FUEL HYDROGEN CONTENT (WT%)

FIGURE 1. EFFECT OF FUEL HYDROGEN CONTENT ON EXHAUST SMOKE FROM ACTUAL ENGINES

fuels with equal hydrogen content (12.8 percent) were blended to stress different molecular components and final boiling points. The fuels containing high concentrations of polycyclic aromatics and partially-saturated polycyclic structures, such as tetralin, produced more soot than would be expected from typical petroleum-based fuels. However, fuels containing naphthenes, such as decalin, had the same sooting tendency as the petroleum-derived fuel. Changes in viscosity and final boiling point did not affect the sooting tendency. Thus, liquid phase pyrolysis was not a significant factor in soot formation. These measurements were made in a Phillips 2-inch research combustor over a wide range of operating conditions. The results showed that the deviation in the sooting tendencies of the fuels containing polycyclic aromatics from that of the petroleum-based fuels varied significantly with operating conditions.

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In a follow-on project (13), the same six test fuels were burned in a T-63 gas turbine combustor at full power, and the sooting tendency was measured in terms of the flame radiation in the primary zone of the combustor. All the test fuels gave essentially the same flame radiation intensities, indicating that soot formed in this combustor was insensitive to the effects of polycyclic aromatics.

Others (14) have also reported varied affects of polycyclic aromatics on soot formation in different combustor designs. The evidence seems to indicate that burner inlet conditions and combustor configuration both play an important role in determining the effects of hydrogen content and polycyclic aromatics on the sooting tendency.

II. OBJECTIVES

The objectives of this experimental program were to:

- Gain a more basic understanding of the role played by fuel composition, i.e., H/C ratio and molecular structure, in the sooting tendency of gas turbine engines.

- Examine the role of flame temperature on the soot concentration in the primary zone of gas turbine combustors.

- Measure the effects of combustor-operating parameters on the sensitivity of sooting tendency to fuel composition, and compare these results with those from full-scale engine tests.

- Compare the sooting tendencies of fuels containing alkyl benzenes with those containing alkyl naphthalenes and tetralins and examine the effects of operating conditions on these fuels.

III. SCOPE

Combustion performance measurements including flame radiation, flame opacity, flame temperature, gaseous emissions, and combustion efficiency were made using a high-pressure research combustor with several test fuels to determine the effects of flame temperature, H/C ratio, and molecular structure on the soot found in the primary zone of the burner. Test fuels were blended with a wide variation in H/C ratio and special emphasis was placed on preparing fuel blends rich in polycyclic aromatics. The effects of combustor operating conditions on the sensitivity of sooting tendency to H/C ratio and polycyclic aromatic components were also examined.

IV. APPROACH

The work was carried out in two phases. In the first phase, the relative effects of H/C ratio and flame temperature on the sooting tendency were measured and compared. Basically, two experiments were performed in this phase. First, the burner inlet temperature was varied from 533K (500°F) to 922K (1200°F) while keeping gas density and mass flow rates of the fuel (Jet A) and the air constant. These tests were repeated for three different stoichiometries. Then the combustor operating conditions were held constant while fuel properties were allowed to vary. For both experiments, the flame temperature was measured optically using the Kurlbaum Method of soot intensity reversal, and the relative soot concentration in the primary zone of the burner was determined using flame opacity.

In the second phase of the work, fourteen test fuels containing different concentrations of alkyl benzenes, methyl naphthalenes, tetralin, and indene, with hydrogen contents ranging from 11.5 to 14.2 percent, were burned in the Phillips 2-inch research combustor over a wide range of operating conditions. Instead of measuring the relative soot concentration by a flame opacity measurement, the sooting tendencies of the fuels were indicated by the relative flame radiation intensity in the primary zone.

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V. EXPERIMENTAL FACILITIES AND METHODS

A. General Description

This work was performed in the combustor facility of the U. S. Army Fuels and Lubricants Research Laboratory. This facility was specially designed to study fuel-related problems in the operation of turbine engines. The air supply system provides a clean, smooth flow of air to the combustion test cell with mass flow rates up to 1.1 kg/s, pressures to 1620 kPa (16 atm), and temperatures to 1086K (1500°F) (unvitiated). Turbine flowmeters and strain-gage pressure transducers are used to measure flow properties of the air and fuel. Thermocouples are referenced to a 339K (150°F) oven. Data reduction is performed on-line with test summaries available immediately; these summaries provide average flow data as well as standard deviations (typically less than 1 percent of average value), exhaust temperatures, profiles, emissions data, and combustion efficiencies.

B. The Phillips 2-Inch Combustor

Figure 2 is a schematic of the Phillips 2-inch combustor. Basically, this combustor is a straight-through cylindrical type, with fuel atomization by a simplex pressure atomizer. The combustor liner is constructed from 2-inch, Schedule 40, Inconel pipe. Film cooling of surfaces exposed to the flame is accomplished by internal deflector rings. The combustor rig is a more rugged version of the original Phillips rig: some major design changes were incorporated for the purpose of reaching higher inlet temperatures 1086K (1500°F), namely uprating the flanges to the 1500-lb class and fabricating them and the combustor housing from 316 stainless steel. Figure 3 shows the combustor installed in the test cell. This combustor has been used extensively in turbine-fuels research for NASA and the Navy in the areas of radiation, smoke, and corrosion research.(15, 16)

Only a limited number of test conditions, mentioned later, were used in Phase I. In Phase II, the burner was operated at 35 test conditions, which were combinations of the nominal air/fuel flow conditions given in Table 1.

RADIATION SENSOR

COMBUSTION
HOUSING

SAPPHIRE
WINDOWS

EXHAUST
PROBES

AIR IN TO
SWIRL PLATE

AIR IN

EXHAUST
GAS OUT

QUENCH AIR

SECONDARY COMBUSTION AIR

PRIMARY COMBUSTION AIR

AIR IN TO SPLASH COOLING RINGS

2 INCH PIPE FLAME TUBE

FUEL IN

FUEL NOZZLE SHIELD
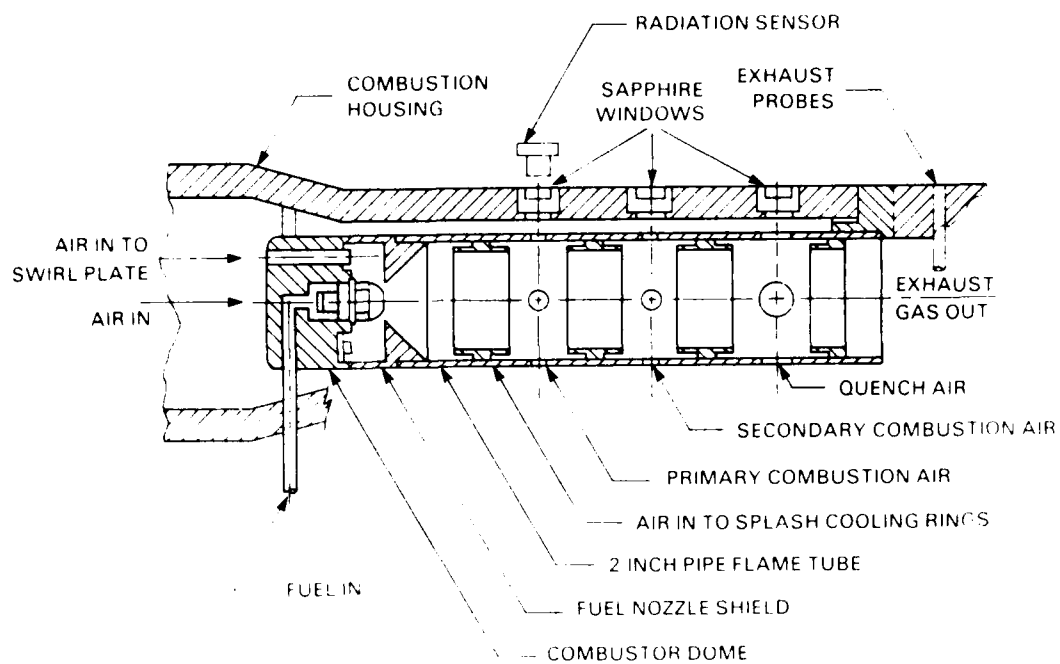
COMBUSTOR DOME

FIGURE 2. PHILLIPS 2-INCH COMBUSTOR
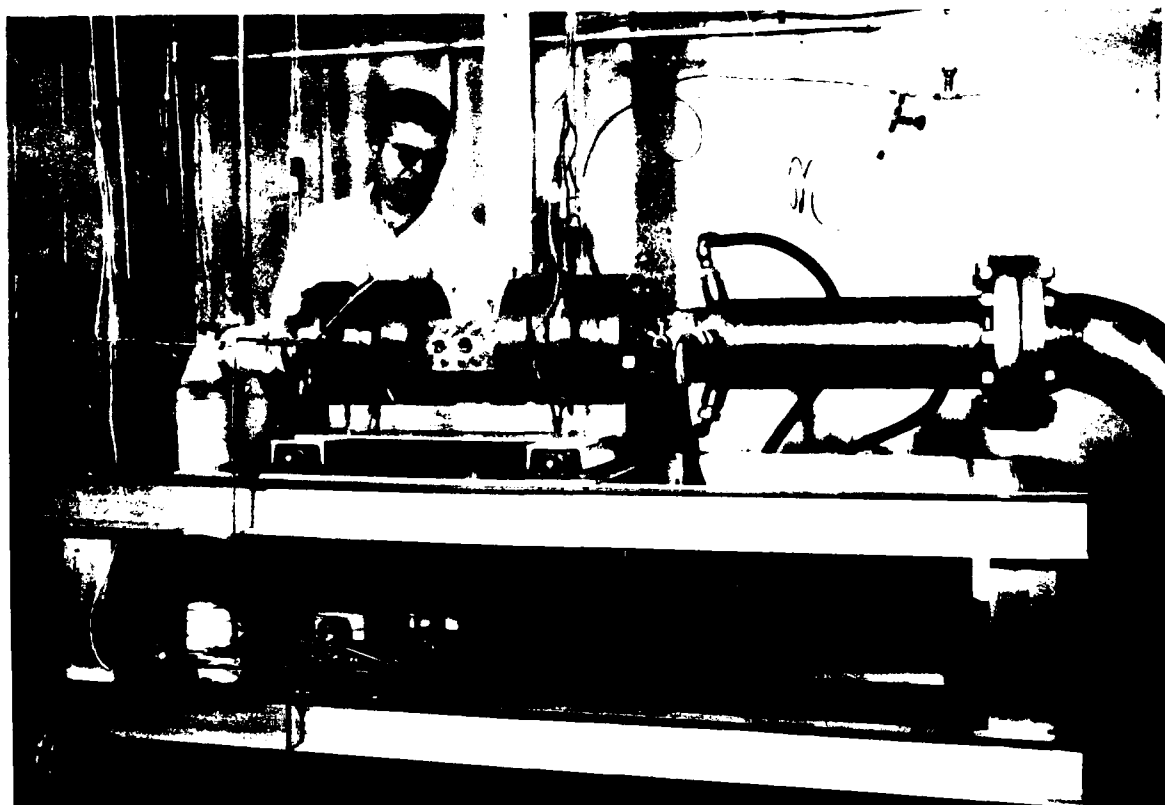
FIGURE 3. VIEW OF COMBUSTOR RIG

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TABLE 1. COMBUSTOR OPERATING CONDITIONS

ρ, kg/m^3	1.64, 2.64, 3.44, 3.29, 3.86, 4.38, 5.2, 5.5, 6.6, 7.7, 8.1
T, K	451, 535, 644, 728, 812, 922, 1031
Reference Velocity, m/sec	18, 30, 60, 90
F/A ratio	0.007, 0.01, 0.0125, 0.015, 0.018

C. The Combustor Facility

A detailed layout of the AFLRL combustor laboratory is shown in Figure 4. The facility consists of a variable pressure-temperature air supply, a control room for operating the air flow system, the fuel flow system, the combustor and its exhaust system; also, there are components for the data acquisition system (see Figures 5 and 6 for views of the control panel).

1. The Air Flow System

A flow diagram of the "air factory" is shown in Figure 7. The compressed air for the lab is generated in two stages: two Ingersoll Rand "Pack-Air" rotary-screw compressors are connected in parallel, each delivering 0.573 m^3/sec (1000 SCFM) at 700 kPa (100 psig). This air goes through an intercooler and then to a single-cylinder reciprocating compressor where it is compressed to 1.75 MPa (250 psia). From there, the air passes through an aftercooler, a receiver, and an oil filter before going to the flow controls. The oil carryover is less than 5 ppm. Suction and discharge bottles are on the booster compressor which, in conjunction with the receiver, were designed on an analog computer by Ingersoll Rand to eliminate pulsations from the air flow. At the downstream side of the receiver, the pressure fluctuation (a frequency of about 45 Hz) was less than 700 Pa (0.1 psi) when the actual pressure was 1.6 MPa (235 psia).

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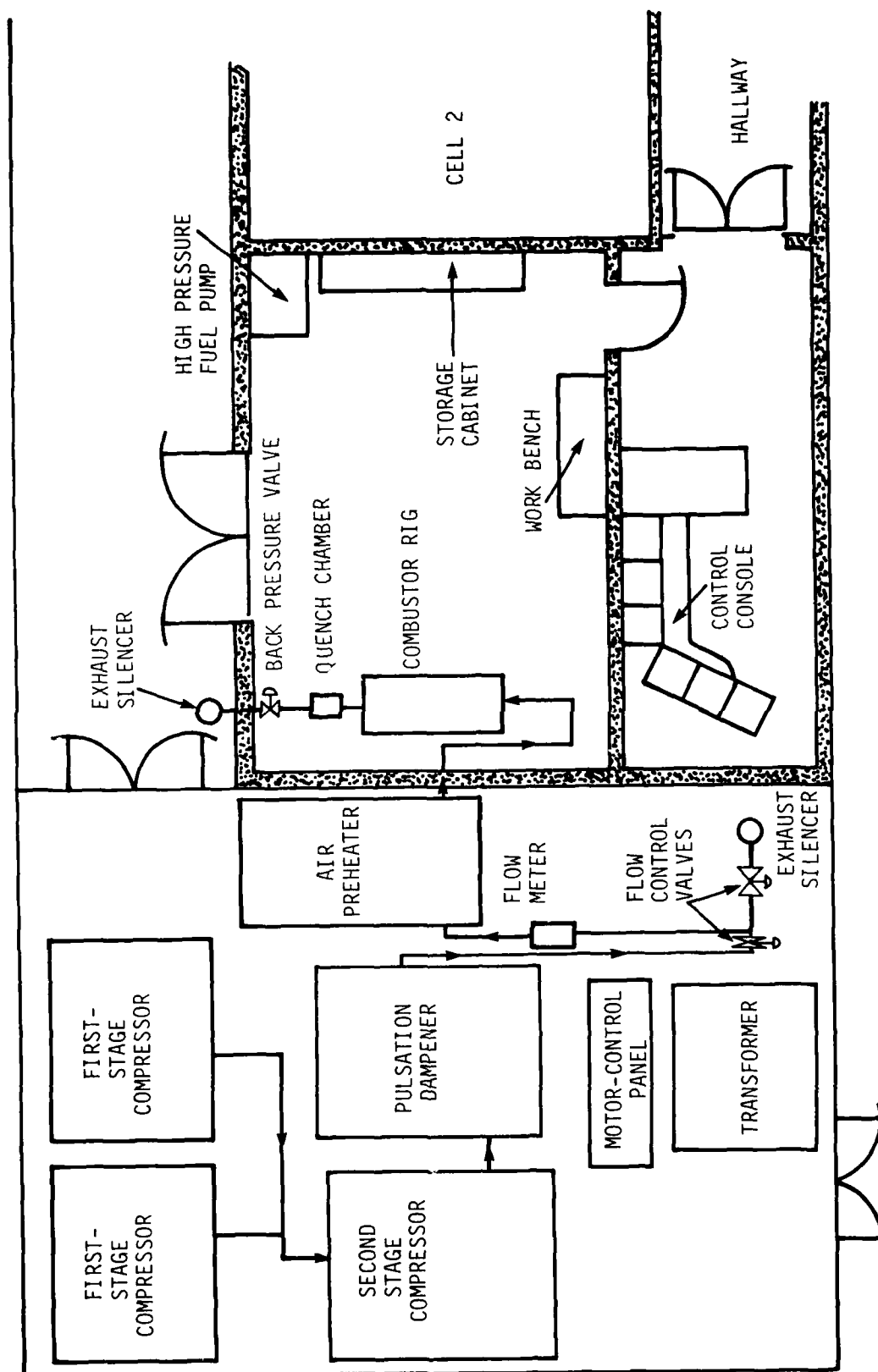
FIGURE 4. LAYOUT OF TURBINE FUEL RESEARCH COMBUSTOR LABORATORY

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a. Air Heater Control System

b. Compressor Motor Controls

c. Pressure Transducer Reference System

d. Thermocouple Reference Oven

e. Moisture Readout

f. Quench Water Control

g. Air Flow Control

h. Ignition and Fuel Flow Control

i. Window Looks Into Combustion Room

FIGURE 5. VIEW OF CONTROL CONSOLE

a. Programmable Calculator

b. Printer

c. X-Y Plotter

d. Scanner and Digital Voltmeter

e. Magnetic Tape Cassette

FIGURE 6. VIEW OF CONTROL CONSOLE SHOWING
DATA ACQUISITION SYSTEM

16
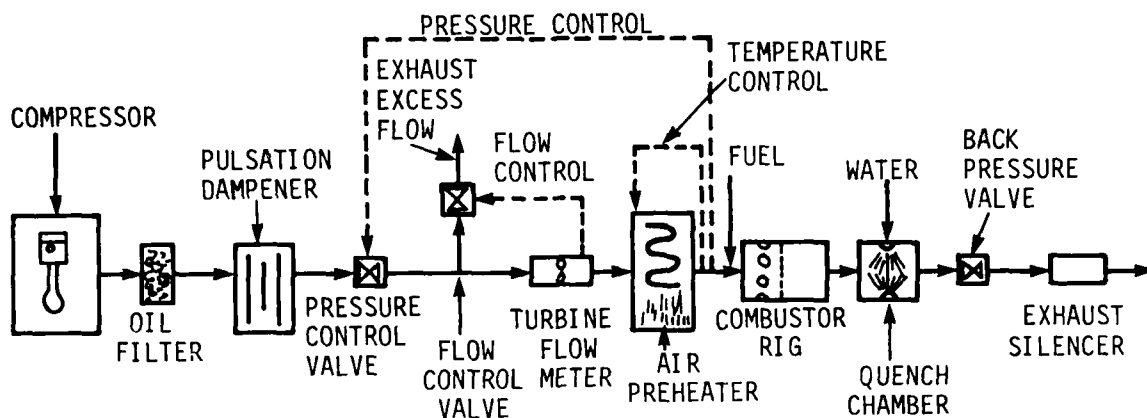
FIGURE 7. FLOW DIAGRAM OF TURBINE COMBUSTOR SYSTEM

The flow control system operates in two parts: one valve is used to provide a pressure drop to the system, while a second valve bypasses any excess air flow through an exhaust silencer. The compressors are always operating at full capacity--a method which uses more total energy but eliminates any surging caused by the compressors unloading.

A 3-inch (7.62-cm) turbine flowmeter is used to measure the air flow rates. Because a turbine meter measures volumetric flow, the pressure and temperature are also sensed at the meter so the flow measurement can be converted to mass flow rate.

The air flow then enters a preheater which is capable of heating the flow from roughly 310K (100°F) to 1116K (1550°F). This heater is an indirect, gas-fired system with a counterflow heat exchanger; the air remains unvitiated. The combustion control system was designed in accordance with federal safety standards. The preheater will shut down automatically in event of a malfunction in the fuel supply or when temperatures exceed established limits. The final air temperature is automatically controlled by a Honeywell recorder-controller system which regulates the air/fuel ratio in the combustion chamber and dilutes the hot exhaust gases going to the tube bundle.

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The air flow is piped into the test cell and, for all practical purposes, is the same as the air from any turbine engine compressor. It is essentially pulsation and oil free, and its moisture content is controlled. The air flow rate, pressure, and temperature are independently adjustable to any value within the operating envelope.

2. The Fuel Supply System

The fuel supply system is capable of pumping fluids ranging in properties from gasoline to No. 5 diesel at flow rates of over 0.063 lb/sec (1 gal./min) and pressures up to 7 MPa (1000 psi). For this program, the fuel was forced from drums to the fuel selection manifold system (see Figure 8) with pressurized inert gas. The manifold employs twelve solenoid valves (for 12 fuels). After the manifold, a high-pressure pump delivers fuel to the combustor. The plumbing from the pump to the combustor is stainless steel to facilitate cleaning when special fuels or fuel additives are used.

A turbine flowmeter measures the flow rate of the fuel. On starting, a system of valves and bypasses is used to increase the flow rate to the desired level before introducing it to the combustion chamber. On shutdown, the lines can be drained and purged with an inert gas.

3. Exhaust System

A pneumatically-controlled valve is located downstream of the quench section to maintain the pressure in the combustor system. A silencer is used to attenuate the exhaust noise.

D. Data Acquisition System

The data acquisition system uses a Hewlett-Packard 9820 programmable calculator with associated hardware. Figure 9 shows a flowchart of the system. A digital voltmeter is coupled to a 50-channel scanner which samples the voltage outputs from the various sensor systems and then feeds the corresponding digital values to the calculator. The calculator handles all of the data reduction and any necessary calculations, e.g., combustion

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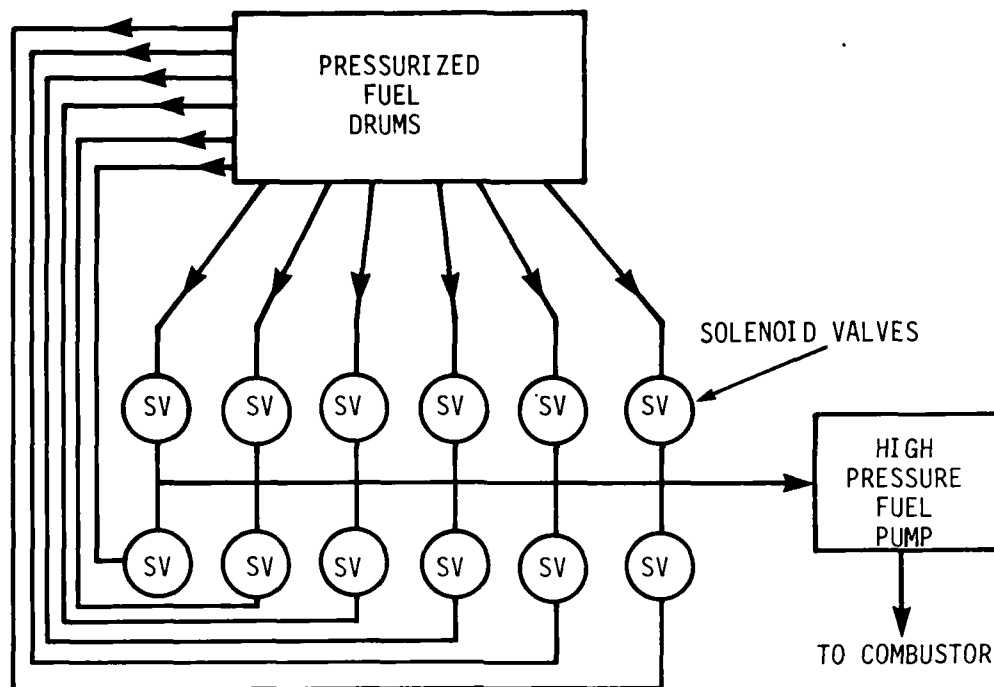
FIGURE 8. FUEL SELECTION MANIFOLDING SYSTEM
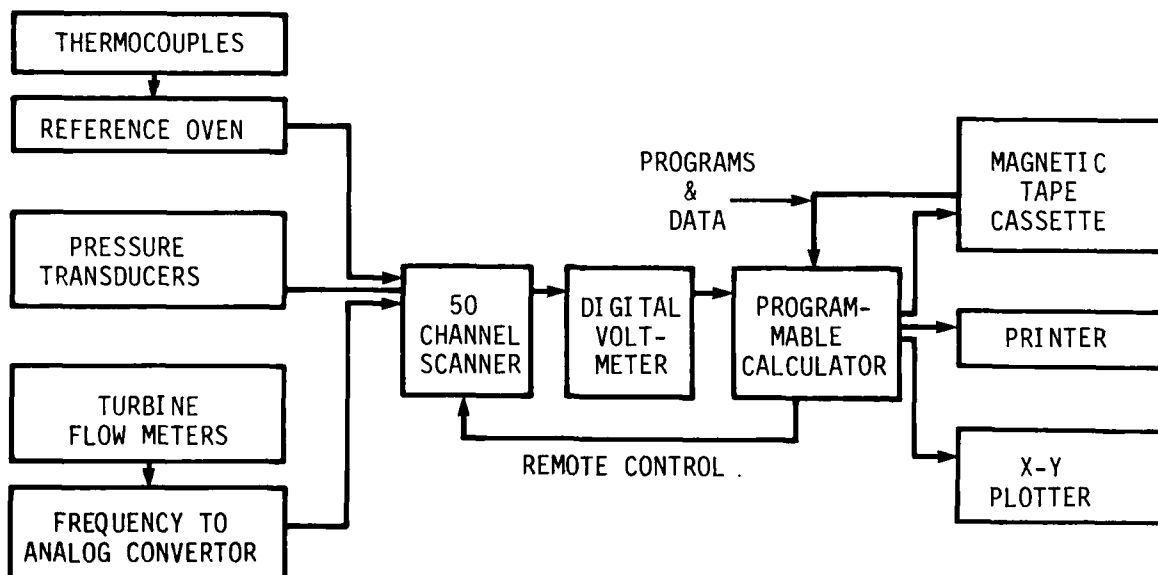
FIGURE 9. DATA ACQUISITION SYSTEM

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efficiency, flow factor, and exhaust emissions coefficients. The resulting data are then processed in one of three ways:

1. The data can be stored on magnetic tape for further reduction at a later time;

2. The data can be output graphically on an X-Y plotter, or

3. The data can be output on a printer along with any appropriate alphanumeric titles or column headings.

Figure 10 shows an example of graphical output. Table 2 illustrates the use of the printer for the continuous monitoring of several channels of data,
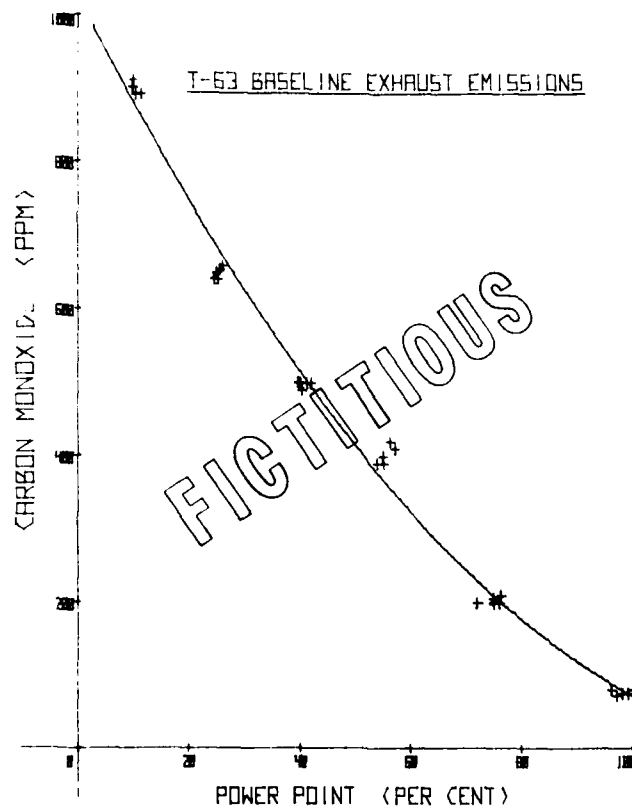
FIGURE 10. EXAMPLE OF GRAPHIC OUTPUT

TABLE 2. EXAMPLES OF CONTINUOUS MONITORING OF
SEVERAL DATA CHANNELS

while Figure 11 is an example of a complete test report available immediately after the data were taken. The 50 channels can be scanned, the data reduced, and a report such as Figure 11 printed out in less than 2 minutes.

The sensing systems consist of strain-gauge pressure transducers, thermocouples, and turbine flowmeters. Regulated power supplies are used with the pressure transducers. A vacuum/pressure reference system is used to calibrate the transducers against a Wallace and Tiernan gauge; use of three-way valves allow this to be done during a test without disconnecting the transducers. The thermocouples are referenced to a 338.5K (150°F) oven; the unit will handle up to 50 thermocouples of any kind, including platinum.

E. Exhaust Analysis Instrumentation

Exhaust emissions are measured on-line using the following instruments in accordance with SAE-ARP 1256 with the exception of the NO/NO_x measurements. These measurements are performed by chemiluminescence.

Sample	Instrument	Sensitivity
Carbon Monoxide	Beckman Model 315B NDIR	50 ppm to 16%
Carbon Dioxide	Beckman Model 315B NDIR	300 ppm to 16%
Unburned Hydro-carbons	Beckman Model 402 FID Hydrocarbon Analyzer	0.5 ppm to 10%
Nitric Oxide	Thermo-Electron 10A Chemiluminescence Analyzer	3 ppm to 10,000 units
Total Oxides of Nitrogen	Thermal-Electron 10A Chemiluminescence Analyzer with NO_x Converter	3 ppm to 10,000 units
Oxygen	Beckman Fieldlab Oxygen Analyzer	0.1 ppm to 100%

The exhaust sample is routed to the instruments through a 177°C (350°F) heated Teflon® line and then appropriately distributed.

FIGURE 11. EXAMPLE OF A TEST REPORT

U.S. ARMY FUEL ... ... RESEARCH LABORATORY
TURBINE COMBUSTOR FACILITY

*** FUEL QUALITY ... ... ANALYSIS ***

... ...

DATE: 10 2 81   TIME: ...
COMBUSTION SYSTEM: ... INCH ... COMBUSTOR
TEST FUEL: JET A + TETRALIN+++

***** EXPERIMENTAL CONDITIONS *****

	AVERAGE	STD DEV.	DESIRED	LAST SCAN
INLET AIR PRESSURE, ...	...	...	.50	115.00
INLET AIR TEMPERATURE, (DEG F)	...	1.41	157.03	100.11
AIR FLOW RATE, LB/SEC	...	...	3.110	.412
FUEL FLOW RATE, KG/MIN	...	...	4.536	.120
FUEL AIR RATIO	...	...	.00002	.00484
REFERENCE VELOCITY ...	...	.4	3.0	31.0
HEAT INPUT	...	...	4.	...5.0

FUEL PRESSURE ...
FUEL TEMPERATURE ...

...

... ...

EXHAUST TEMPERATURE ...

... ...
... ...
... ...
... ...

...

... ...
FT ...
NO ...
54. ...

SMOKE NUMBER:

COMBUSTION EFFICIENCY ...
FUEL AIR RATIO (CALCULATED FROM) ...   .000...
REMARKS:

FIGURE 11. EXAMPLE OF A TEST REPORT

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F. Optical Temperature Measurements

Combustors have large temperature gradients in their primary zones where cool air violently mixes with the fuel spray and hot combustion gases. It was important in Phase I of this program to measure a temperature characteristic of those parts of the flame where soot was being formed. The Kurlbaum technique, or soot reversal method, is an ideal way to indicate a flame temperature because it measures the temperature of soot particles in the flame zone. The measured temperature is heavily weighted toward the maximum flame temperature because of the wavelength selected and Planck's radiation law. It is also a well-established technique and straightforward to calibrate, requiring much simpler apparatus than other nonintrusive optical techniques such as coherent antistokes Raman spectroscopy (CARS). However, it is similar to sodium D-line-reversal except that broader band soot emission and absorption are used rather than a spectral line. The Kurlbaum method is described in Gaydon and Wolfhard (17), and the details of the mathematics are presented by Tourin (18). Precautions such as those suggested by Thomas (19) for line-reversal methods have been observed.

The optical setup used in the Kurlbaum method is depicted in Figure 12. The dependence of detector signal on lamp current is first measured without the flame in the optical path; then the same measurements are repeated with the flame present. The region where these two plots of signal versus lamp current cross is the point of reversal. The spectral brightness temperature of the tungsten strip lamp at the reversal point is equal to the flame temperature after allowing for window and lens losses on the lamp side, and correcting the brightness temperature for the difference in pyrometer wavelength ($\lambda = 0.65\,\mu m$) and flame measurement wavelength ($\lambda = 0.5\,\mu m$). A silicon photodiode with a $0.5\text{-}\mu m$ filter was used as a detector, and a lock-in amplifier was used to amplify the chopped signal. Careful use of apertures, as suggested by Thomas (19), was necessary to assure proper calibration.

G. Opacity Measurement

Flame opacity measurements are used to determine the relative soot concen-

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TUNGSTEN STRIP LAMP

CHOPPER (OPACITY MEASUREMENT)

LENS

AIR FLOW

FUEL FLOW

AIR FLOW

EXHAUST

APERTURE

LENS

CHOPPER (REVERSAL MEASUREMENT)

0.5-..m FILTER (REVERSAL MEASUREMENT)
0.9-..m FILTER (OPACITY MEASUREMENT)

Si DETECTOR
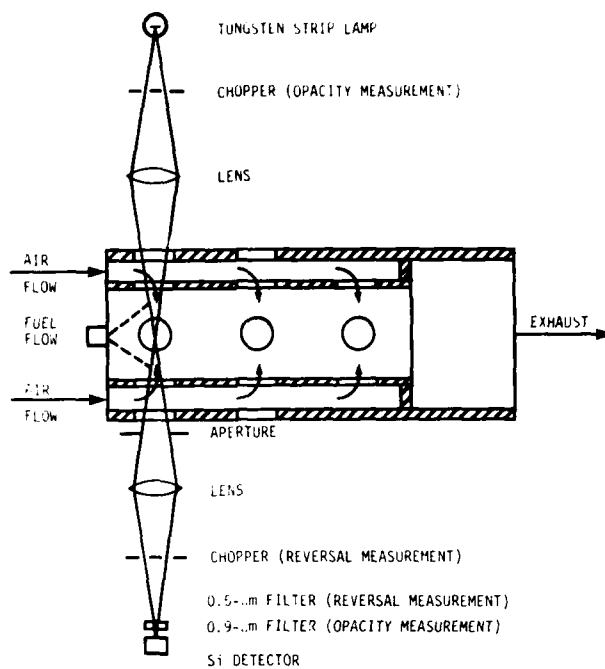
FIGURE 12. EXPERIMENTAL APPARATUS

trations. The same optical apparatus as the above was used for opacity measurements except that the chopper was moved to the lamp side of the combustor to remove flame radiation from the signal, and a 0.9 μm bandpass filter was used to discriminate against absorption not related to soot particles.

H. Radiation Measurement

Flame radiation measurements are used to determine relative sooting tendencies. Flame radiation was measured with a Rayotube, Model 8890-S, detector. A transverse observation of flame radiation was made via sapphire windows mounted in the sides of the combustor housing (see Figure 2).

I. Combustion Efficiency

Combustion efficiencies were calculated from the exhaust gas analysis ac-

cording to a relationship developed by Hardin ([20]):

$$\epsilon = \left[1 - \frac{A \cdot f(UBH) - 121{,}745 \cdot f(CO) - 38{,}880 \cdot f(NO) - 14{,}654 \cdot f(NO_2)}{A \cdot [f(CO_2) + f(CO) + f(UBH)]} \right] 100\%$$

where $f(i)$ is the concentration of "i" in the exhaust, A is a constant based on the heat of combustion and hydrogen/carbon ratio of the fuel, and UBH is unburned hydrocarbons.

J.   Test Fuels

The test fuels were blended by adding a mixture of alkyl benzene (BTX), liquid methyl naphthalenes (LMN), tetralin, and indene in various concentrations to reference base fuels, which were low aromatic kerosenes. BTX, a mixture of aikyl benzenes, was obtained from a local refinery. The LMN was obtained from another company and consisted of about 80 percent alkyl naphthalenes, with the remainder being naphthalene and biphenyl. Tetralin solvent, at least 97 percent pure, was obtained from one company, and the indene came from still another company. Two reference fuels were used during the course of this work because the one in use at the start of the project was depleted midway; however, a low aromatic kerosene (Jet A-1) with essentially the same hydrogen content as the original reference fuel Jet A was found. The only significant difference in these fuels was the distribution of single and double ring aromatics; the Jet A-1 had a higher concentration of double-ring aromatics. When the two reference fuels were burned in the Phillips research combustor at several operating conditions, a difference in sooting tendency could not be determined. A description of the test fuels including the reference fuels is given in Table 3, and a summary of the properties of the reference fuels and blending materials is shown in Table 4.

The second base fuel, Jet A-1, was used only in blending the test fuels containing tetralin and indene and was used as a reference fuel when these fuels were tested.

TABLE 3. TEST FUEL DESCRIPTIONS

Fuel No.	Composition	Hydrogen Content, wt%	H/C Ratio
0	Jet A	14.2	1.971
1	Jet A + 17.0% BTX	1.35	1.859
2	Jet A-1 + 12.8% Tetralin	13.5	1.859
3	Jet A + 11.2% LMN	13.5	1.859
4	Jet A + 42.0% BTX	12.5	1.702
5	Jet A + 21.0% BTX + 12.7% LMN	12.5	1.702
6	Jet A-1 + 32.6% Tetralin	12.5	1.702
7	Jet A + 25.5% LMN	12.5	1.702
8	Jet A + 65.5% BTX	11.5	1.548
9	Jet A + 43.0% BTX + 13.6% LMN	11.5	1.548
10	Jet A + 22.0% BTX + 26.4% LMN	11.5	1.548
11	Jet A + 39.7% LMN	11.5	1.548
12	Jet A-1 + 27.4% BTX + 30.4% Tetralin	11.5	1.548
13	Jet A-1 + 52.3% Tetralin	11.5	1.548
14	Jet A-1 + 36.55% Indene	11.5	1.548

TABLE 4. PROPERTIES OF REFERENCE FUELS AND BLENDING AGENTS

Fuel Type	Jet A	Jet A-1	BTX	LMN	Tetralin
Heat of Combustion Net (J/g)	43,575	43,899	40,679	38,512	41,003
Carbon, wt%	85.78	85.78	89.8	91.89	90.92*
Hydrogen, wt%	14.2	14.15	10.1	7.26	9.08*
Aromatics by UV					
Single ring, wt%	4.6	2.50	52.5	7.2	54.54*
Double ring, wt%	1.3	4.7	1.7	75.2	---
Triple ring, wt%	0.02	0.02	0.01	1.4	---
Total, wt%	5.92	7.22	54.21	83.8	54.54
Aromatics by FIA					
Aromatics, vol%			8.53	97.1	
Olefins, vol%			0.81	1.4	
Saturates, vol%			90.66	1.5	
HPLC Analysis					
Saturates, wt%	91.9				
Aromatics, wt%	8.1				
Viscosity @ 313K, cSt	1.68	1.72			
Specific Gravity	0.801	0.8049	0.8702	1.0173	0.970

*Calculated

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VI. RESULTS AND DISCUSSION (PHASE I)

Two basic experiments were performed to determine the effects of temperature and fuel hydrogen content on soot formation. The purpose of the first experiment was to determine the effect of flame temperature on soot concentration without any variation in fuel properties; in the second experiment, the fuel properties were varied and the operating conditions were held constant. The burner inlet temperature was varied from 533K to 922K (500°F to 1200°F), while the gas density, reference fuel (Jet A) mass flow rates, and inlet air mass flow rate were held constant. Table 5 shows the burner inlet temperatures and corresponding burner inlet pressures required to maintain constant gas density for the six operating conditions examined. These tests were repeated for three different overall stoichiometries, i.e., fuel/air ratios of 0.0096, 0.0108, and 0.0136. The fuel/air ratios in the primary zone of the combustor were approximately 0.029, 0.032, and 0.041 because it is assumed only one third of the air enters this zone. The

TABLE 5. COMBUSTOR CONDITIONS

Operating Condition	1	2	3	4	5	6
Burner Inlet Temperature, K	533	589	644	728	812	922
(°F)	500	600	700	850	1000	1200
Burner Inlet Pressure, kPa	786	862	938	1082	1200	1344
(psia)	114	125	136	157	174	195

Reference velocity and burner inlet gas density were held constant
at 30.5 m/sec and 5.26 kg/m^3, respectively.

flame temperature (soot temperature by the Kurlbaum technique) and the relative soot concentration (flame opacity) were measured in the primary zone of the combustor at each operating condition. Figure 13 shows the effect of burner inlet temperature on flame temperature. (Lines through the data are based on a least-squares fit for all figures.) The change in flame temperature with burner inlet temperature is in good agreement with adiabatic flame temperature calculations. An increase in burner inlet temperature from 533K to 922K (500°F to 1200°F) changes the calculated stoichiometric adiabatic flame temperature from 2400K to 2590K (3860°F to 4203°F) or

7.9 percent. The measured flame temperatures increase from about 2140K to 2297K (3393°F to 3675°F) or 7.3 percent. It seems reasonable that convective and radiative heat losses would account for the 200 to 300K difference between the measured temperature and the stoichiometric adiabatic flame temperature. The measured flame temperatures actually decreased slightly instead of increasing dramatically with fuel/air ratio as is observed in premixed flames.(17) The decrease in flame temperature with increasing fuel/air ratio may have been caused by a change in flame position; when the differential fuel pressure across the atomizing nozzle is increased, the fuel spray has greater penetration, and the flame is expanded further into the flame tube.

The effect of flame temperature on the relative soot concentration (flame opacity) is shown in Figure 14. Clearly, the soot concentration increases with both increased flame temperature or inlet temperature and increased fuel/air ratio.

The flame temperature measurements and the soot concentration measurements are consistent with diffusion flame behavior, i.e., the flame zone temperature where soot is formed is near the stoichiometric flame temperature and the soot concentration increases as the flame temperature increases. The dependence of soot concentration on temperature is shown by the Arrhenius correlation in Figure 15. In this analysis, the relative soot concentration is assumed to be proportional to the global rate of soot formation and oxidation in the primary zone of the burner. All three flame stoichiometries in Figure 15 show a very similar temperature dependence. The apparent activation energies from the leanest to the richest condition were 42 kcal/mole, 47 kcal/more, and 55 kcal/mole. These values are not unreasonable when compared to those obtained by Millikan and others (10) for the activation energy of soot production from acetylene, 34, 39, 47, 50 and 38 kcal/mole. The activation energies obtained by Glassman and Yaccarino (9) for soot formation in diffusion flames of several compounds range from 85 to 115 kcal/mole. It appears that their method of measuring the temperature dependence of soot formation emphasizes the pyrolysis reactions since the activation energies are comparable with C-C and C-H bond energies in hydrocarbon molecules.

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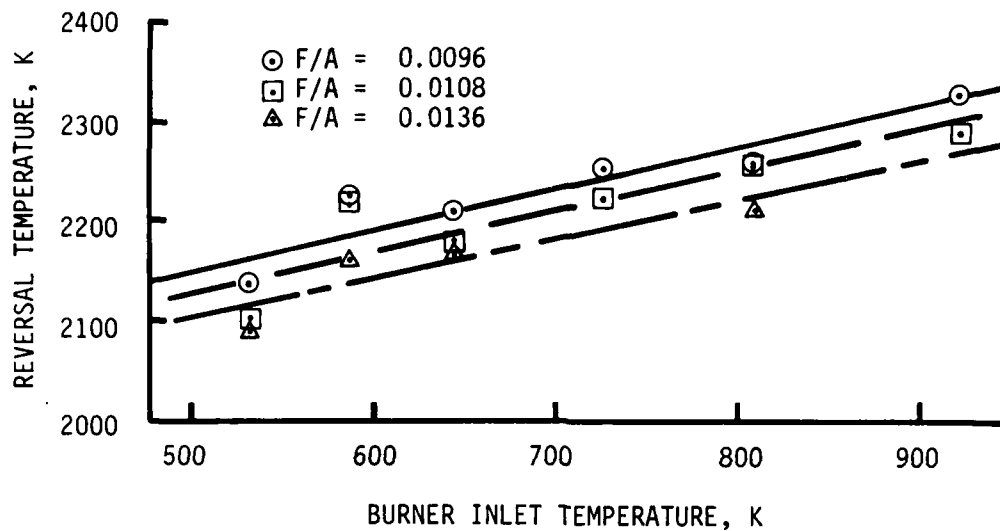
FIGURE 13. THE EFFECT OF BURNER INLET TEMPERATURE
ON FLAME TEMPERATURE AS MEASURED BY THE KURLBAUM TECHNIQUE
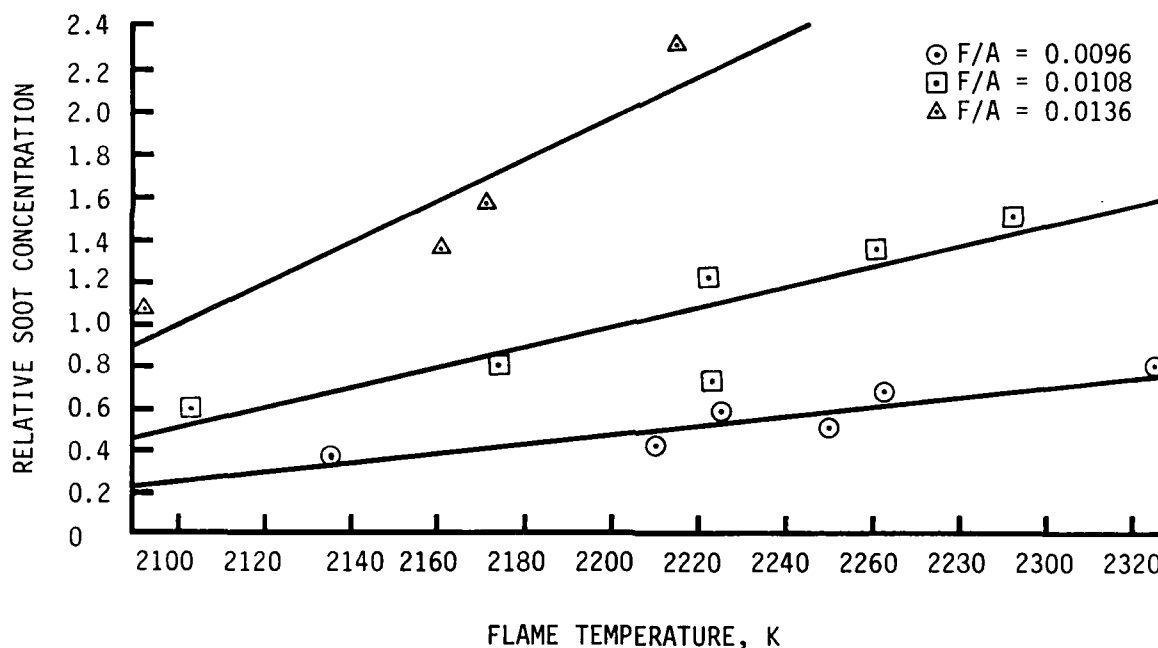
FIGURE 14. THE EFFECT OF FLAME TEMPERATURE ON
SOOT CONCENTRATION

The purpose of the second experiment was to determine the effect of H/C
ratio on flame temperature and soot formation. Ten test fuels were burned
at operating condition number 3 (see Table 5) at a fuel/air ratio of 0.0098.

30
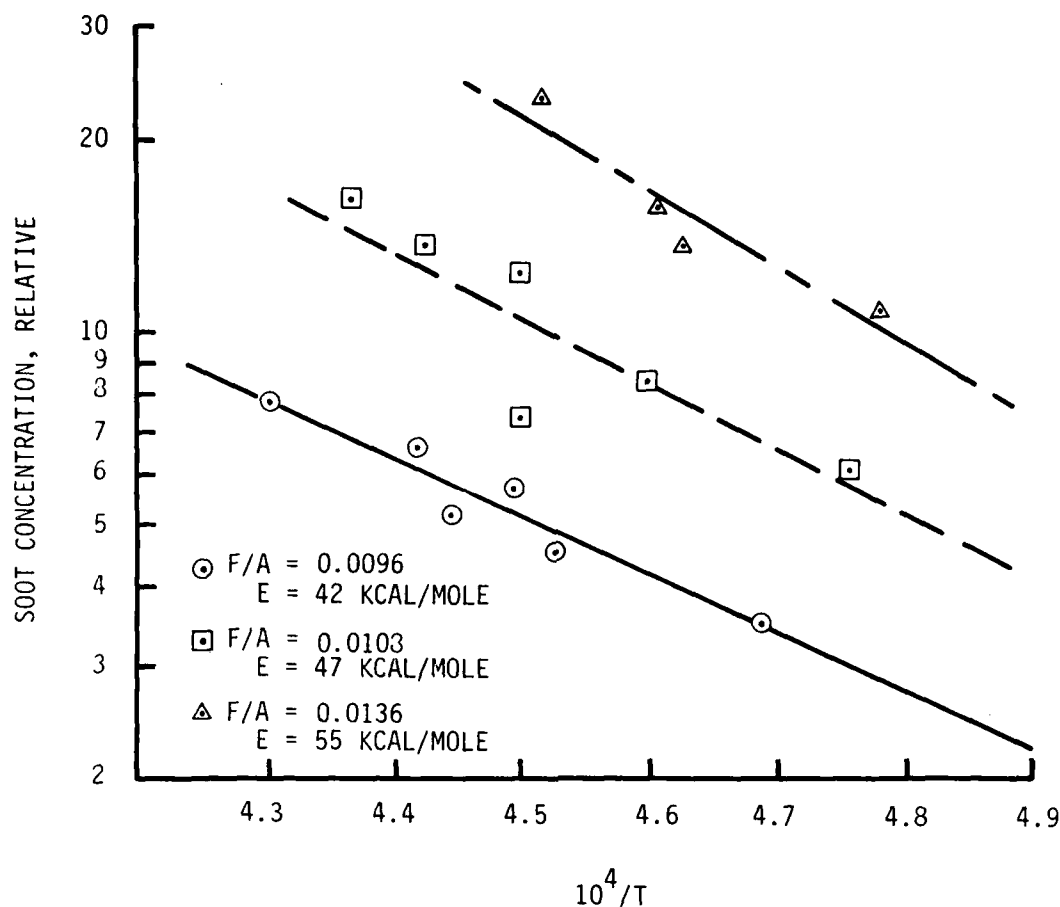
FIGURE 15. ARRHENIUS CORRELATION OF SOOT CONCENTRATION AND
FLAME TEMPERATURE FOR THE REFERENCE FUEL JET A

The test fuels number 0, 1, 3, 4, 5, 7, 8, 9, 10, and 11 were a blend of the reference fuel Jet A, high-boiling alkyl benzenes (BTX), and liquid methyl naphthalenes (LMN). These fuels had been originally blended for Phase II of the program to study the effects of polycyclic aromatics on soot formation, but their wide range of hydrogen content (11.5 to 14.2 percent) rendered them suitable for this experiment.

Figure 16 shows that the flame temperature decreased as the hydrogen/carbon ratio is increased, but the actual magnitude of the change in flame temperature is small when compared with the observations from the first experiment involving changes in burner inlet temperature (See Figure 13). For the range of H/C ratios from 1.55 to 1.98, the flame temperature changes by only 35K while the soot increases by a factor of about 2.5. If the change in

31
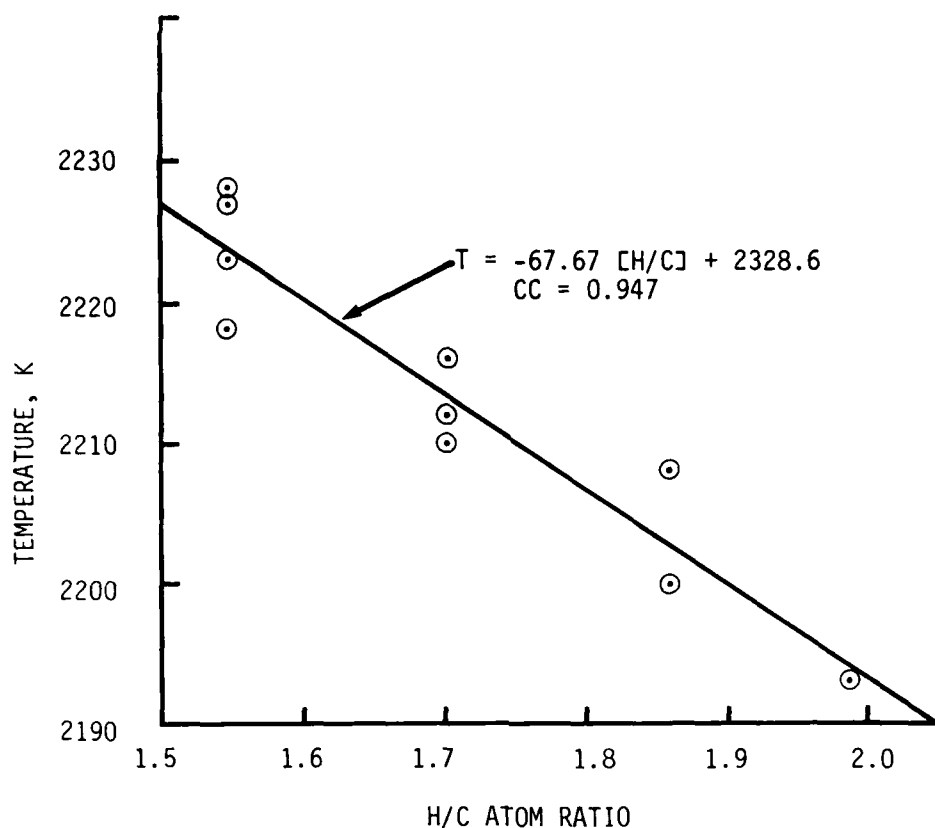
FIGURE 16. THE EFFECT OF H/C ATOM RATIO OF FLAME TEMPERATURE
MEASURED BY THE KURLBAUM TECHNIQUE

soot concentration were attributed solely to changes in flame temperature, formation of soot would be extremely temperature dependent. The Arrhenius correlation of the soot concentration and the measured flame temperature shown in Figure 17 indicate a very high temperature dependence, equivalent to an activation energy of 275 kcal/mole, which is much greater than the 50 kcal/mole measured in the first experiment on temperature effects alone. It is evident that the relatively small changes in flame temperature due to the fuel composition (H/C ratio) have only a slight impact on the propensity of a fuel to soot, and that other chemical effects related to the H/C ratio must be involved in soot formation.
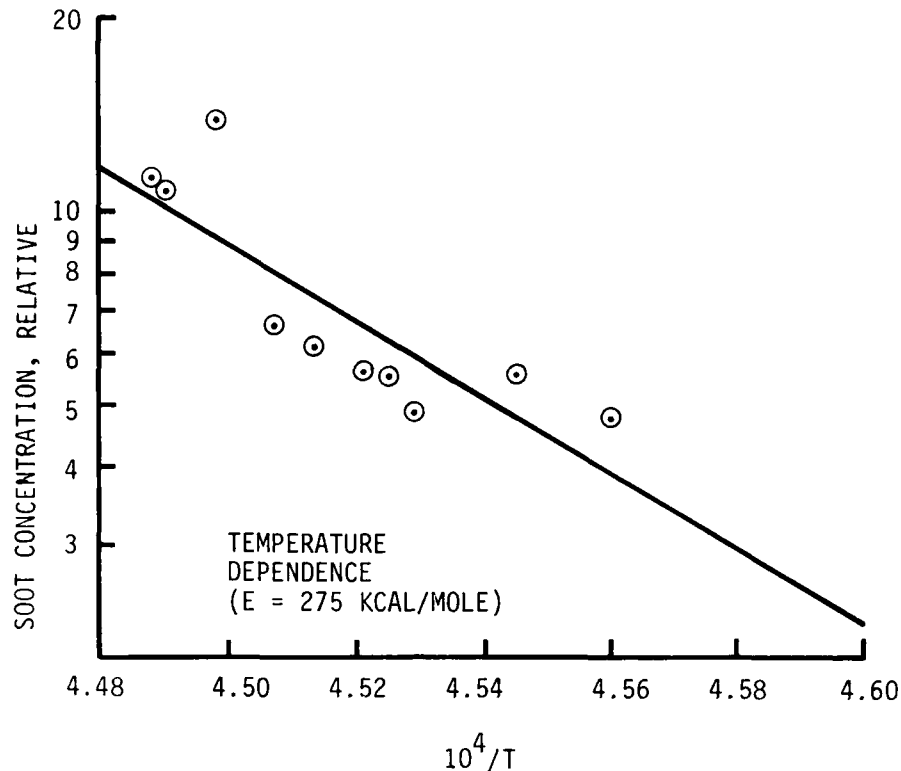
FIGURE 17. ARRHENIUS CORRELATION OF SOOT CONCENTRATION AND
FLAME TEMPERATURE FOR FUELS OF VARIOUS HYDROGEN CONTENTS

VII. EXPERIMENTAL RESULTS (PHASE II)

A. Sooting Tendency

The sooting tendency in gas turbine engines is generally indicated by ex-
haust smoke emissions and increased combustion chamber liner temperature,
i.e., radiant heat transfer from incandescent carbon particles (21, 22).
However, the soot concentration in the primary zone of the burner where soot
is formed can be measured more directly in terms of flame absorptivity. If
flame temperature were independent of fuel composition and the flames were
optically thin (absorptivity ≤ 0.4), then the flame radiation intensity would
be a good relative measure of the sooting tendency of the test fuels. The
results of Phase II showed that flame temperature, i.e., soot temperature,
as measured by the Kurlbaum technique, varied by less than 2 percent for
fuels with hydrogen contents ranging from 11.5 to 14.2 percent. Because

these variations in flame temperature with fuel composition were small, the error incurred by neglecting the temperature differences (Stephen-Boltzmann Law) in relating the soot concentration to the radiation intensity was less than 7 percent. Thus, the flame radiation was considered to be an adequate indicator of the sooting tendency in this study.

B. Hydrogen Content Correlation

It is well known that the flame radiation, R, in gas turbine combustors correlates with the H/C ratio of the fuel (see Introduction). The correlation is generally found to be a linear function of H/C ratio if the range of fuel hydrogen contents is not too large with the slope, dR/d(H/C), of the line usually defined as the sensitivity to hydrogen content(3). However, it is often more useful to compare the relative, or percentage, change in radiation with H/C ratio by using a normalized sensitivity given by:

$$S = \frac{1}{R}\left[dR/d(H/C)\right]$$

This factor, which is analogous to Jackson and Blazowski's(2) normalized liner temperature parameter, can be used to compare the relative sensitivities of different engines or the same engine at different operating conditions. However, this sensitivity turns out to be the slope of the line that results when ln(R) is plotted against H/C ratio. Such a correlation could be tested for relevance only if flame radiation data were available over a wide range of H/C ratios for a particular operating condition. Earlier work on the effects of fuel properties on flame radiation and exhaust smoke in a T-63 combustor for the U. S. Navy (4) and the U. S. Air Force (7) has resulted in a data bank that includes fuels with H/C ratios ranging from about 1.5 to 2.5. These test fuels, which varied significantly in composition,

$\ln R = 8.330 - 1.821(H/C)$
CC = 0.934

FIGURE 18. CORRELATION OF FLAME RADIATION WITH H/C RATIO IN A T-63 GAS TURBINE COMBUSTOR

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included syncrudes, petroleum-base, water-in-fuel macroemulsions, microe-mulsions of water-in-fuel and alcohols-in-fuel, and alcohol-fuel solutions. The radiation intensities of these fuels measured at the full power opera-ting condition of the T-63 combustor were found to correlate favorably with H/C ratio. However, this plot of R versus H/C ratio was not a straight line; instead, it appeared to resemble an exponential dependence of R on H/C ratio. Figure 18 shows that an exponential model gives a surprisingly good correlation (cc=0.934) of ln(R) and H/C ratio. This method of correlating R with H/C ratio is used in the present study because it correlates well with the data and gives a normalized sensitivity to H/C ratio.

C. Sensitivity to H/C Ratio

In analyzing the experimental results, the three sensitivities to H/C ratio given below were used to describe the behavior of fuels blended with mono-cyclic aromatics, blends with tetralin, and blends with polycyclic aroma-tics. As noted above, the sensitivity to H/C ratio to be used in this analysis is defined as $S = 1/R \, [dR/d(H/C)]$.

S_1 - based on fuels 0,1,4,8; Jet A + monocyclic aromatics

S_2 - based on fuels 0,2,6,13; Jet A-1 + tetralin

S_3 - based on fuels 0,3,7,11; Jet A + polycyclic aromatics

The values of S_1, S_2, and S_3 were derived from separate correlations of ln(R) with H/C ratio using the respective fuels designated above. Figure 19 shows a typical data correlation used to determine the values of S_1 and S_3. The data points for the fuels containing tetralin are not on this plot because they nearly always coincided with the data points for the fuels containing polycyclic aromatics. This is shown in Figure 20 where S_2 is plotted against S_3 for several operating conditions. A least squares straight line fit of the (S_2, S_3) points gives a line that falls essentially on the diagonal showing that S_2 equaled S_3 over the range of operating conditions examined. Figure 21 shows a very different result when S_3 is plotted against S_1. It is clear that most of these points deviate signifi-cantly from the diagonal, indicating that the fuels blended with polycyclic
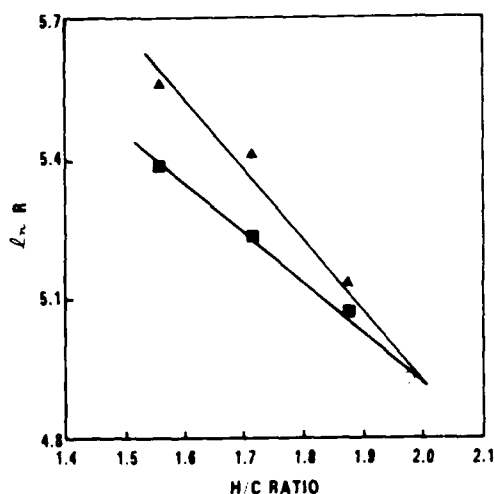
FIGURE 19. CORRELATIONS OF
FLAME RADIATION WITH H/C RATIO
FROM THE PHILLIPS RESEARCH
COMBUSTOR, FUELS 0, 3, 7, 11, ▲,
FUELS 0, 1, 4, 8,■

aromatics and tetralin, as a whole, have a higher sooting tendency than fuels blended with monocyclic aromatics. It is also evident from the number of points which deviate from the diagonal in Figure 21 that the operating conditions have a dramatic effect on the magnitudes of S_3 and S_1.

The effects of a single operating parameter on S_1 and S_3 were evident by simple inspection of the data. For example, the effects of burner inlet temperature were investigated in a series of experiments keeping the gas density, F/A ratio, and reference velocity constant, and using the same atomizing fuel spray nozzle. It was realized that the effect of the fuel spray nozzle may be an important parameter in relating all of the operating conditions. Therefore, with the aid of a Malvern Model 2200 particle sizer, the atomizing nozzles were selected to minimize effects of spray penetration and droplet size.

To correlate S_1 and S_3 with the operating parameters, T, ρ, F/A ratio, and V, all the data were analyzed by a multiple variable linear regression technique. The polynomial functions for S_1 and S_3 given below were found to give a reasonable fit of the data with correlation coefficients of 0.853 and 0.846, respectively.

A polynomial model of the type

$$S = I + a_1T + b_1T^2 + a_2V + b_2V^2 + a_3(F/A) + b_3(F/A)^2 + a_4\rho + b_4\rho^2 \qquad (1)$$

was used in the multiple-variable linear-regression analysis of the data. Values for the constants are given on page 38.

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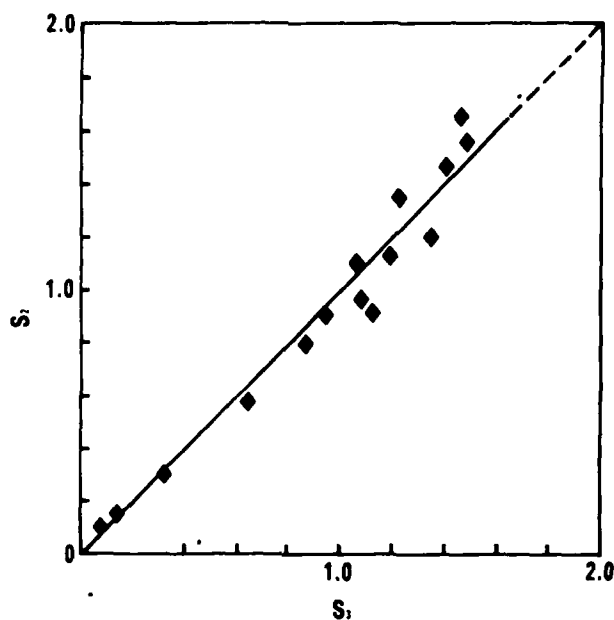
FIGURE 20. COMPARISON OF THE SENSITIVITIES OF THE SOOTING TENDENCY
TO H/C RATIO FOR FUEL BLENDS CONTAINING POLYCYCLIC AROMATICS
(S_3) AND FUEL BLENDS CONTAINING TETRALIN (S_2)
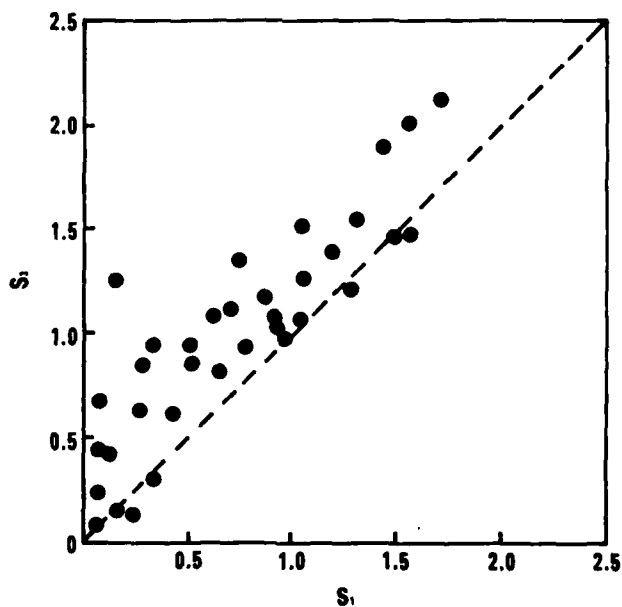
FIGURE 21. COMPARISON OF THE SENSITIVITIES OF THE
SOOTING TENDENCY TO H/C RATIO FOR FUEL BLENDS CONTAINING
POLYCYCLIC AROMATICS (S_3) AND FUEL BLENDS CONTAINING
MONOCYCLIC AROMATICS (S_1)

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Constant	S_1	S_3
I	4.318	2.089
a_1	-1.203	-0.753
b_1	0.062	0.029
a_2	0.021	0.039
b_2	-9.0×10^{-5}	-1.8×10^{-4}
a_3	202.81	170.66
b_3	-6909.13	-7055.18
a_4	-0.138	0.117
b_4	0.011	-0.013

The results are illustrated in Figures 22 and 23. Figure 24 shows the value S_3-S_1, i.e., the increase in sensitivity caused by the polycyclic aromatic components. Each trend line in Figures 22 to 24 expresses the change in that parameter when all other variables are constant. For example, the trend line for burner inlet gas density, ρ, is plotted at constant T, V, and F/A ratio. Pressure is not shown because it would have the same trend as density.

The correlations show that the sensitivities decrease rapidly with increased burner inlet temperature; S_1 decreases faster than S_3. The sensitivities increase rapidly with reference velocity; S_3 increased faster than S_1. Actually, in looking at the raw data, the sensitivities were essentially zero for most of the operating conditions with reference velocities less than 30 m/sec. The sensitivities were affected less by F/A ratio, and changes in density gave the smallest changes.

The differences between S_3 and S_1 are used to express the deviations observed between the fuels blended with polycyclic aromatics and those blended by adding monocyclic aromatics. The analysis shows rather modest variation in S_3-S_1 with temperature, indicating that there is a peak in the sensitivity near 700K and a rapid fall-off at the very high temperatures. The decrease in S_3-S_1 with F/A ratio is quite dramatic and was highly evident in the course of carrying out the experiments. The same was true for the pronounced increase in S_3-S_1 with reference velocity. The trend line for density indicates that S_3-S_1 would be highest at about 5.5 ·kg/m³ and would decrease rapidly at densities above and below that value.
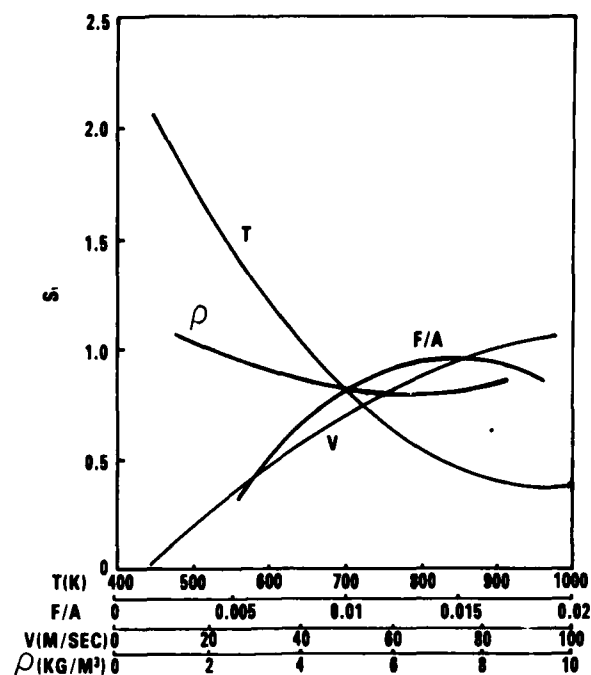
FIGURE 22. EFFECTS OF OPERATING PARAMETERS ON THE SENSITIVITY
OF THE SOOTING TENDENCY TO H/C RATIO FOR FUELS
CONTAINING MONOCYCLIC AROMATICS
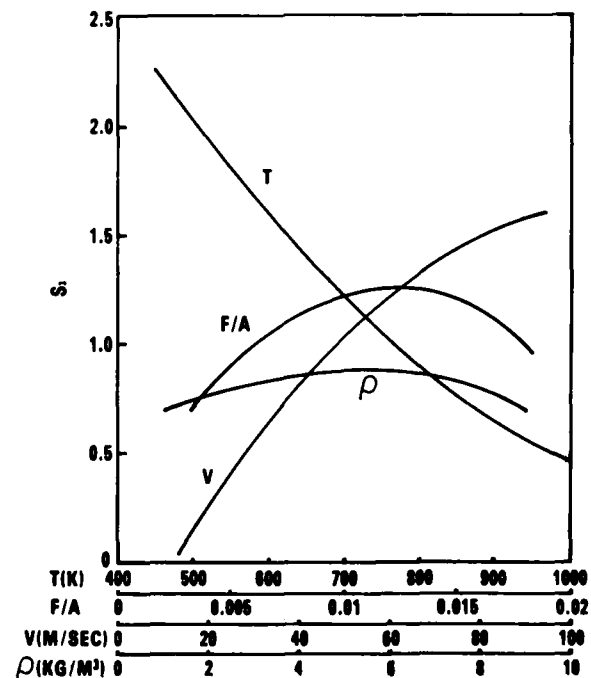
FIGURE 23. EFFECTS OF OPERATING PARAMETERS ON THE
SENSITIVITY OF THE SOOTING TENDENCY TO H/C RATIO FOR
FUELS CONTAINING POLYCYCLIC AROMATICS
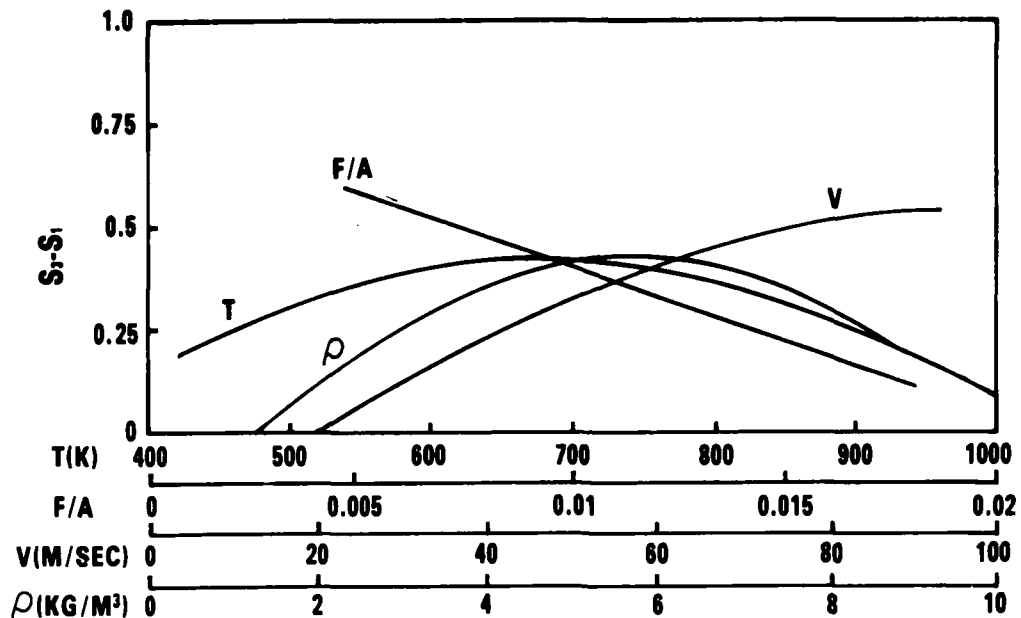
FIGURE 24. EFFECTS OF OPERATING PARAMETERS ON THE
INCREASED SENSITIVITY OF THE SOOTING TENDENCY
DUE TO POLYCYCLIC AROMATICS

D. Concentration Dependence of Polycyclic Aromatics

Fuel blends with hydrogen contents of 13.5, 12.5, and 11.5 percent were
prepared with different concentrations of polycyclic aromatics (see Table
3). For fuels of the same hydrogen content, it was possible to determine
the dependence of flame radiation on the concentration of polycyclic aroma-
tics. Figure 25 shows what was typically observed for the effect of polycy-
clic aromatic ring carbon on the flame radiation. It is apparent that the
curves for the fuel blends containing 12.5 and 11.5 percent hydrogen show a
linear dependence. A linear dependence was also found for the effect of
tetralin ring carbon on flame radiation based on Fuels 8, 12, and 13 con-
taining 11.5 percent hydrogen. It can be seen that raising the polycyclic
aromatic ring carbon to about 25 percent is equivalent to lowering the
hydrogen content by as much as one percent.

E. Indene

Indene is structurally similar to tetralin, except that it has a five-membered side chain containing an olefinic bond. Unlike tetralin, the pyrolysis of indene would probably not form a naphthalene-type intermediate (23), which appears to be the reason why tetralin has a higher sooting tendency than other single-ring aromatics. Fuel No. 14 blended by adding indene to Jet A-1 was only tested at a few operating conditions because of the scarcity and high cost of indene. The tests showed that for the same hydrogen content, Fuel 14 containing indene produced less soot than Fuels 11 and 13 containing polycyclic aromatics, but formed more soot than Fuel 8 containing monocyclic aromatics. An analysis of three runs where there was high sensitivity to molecular structure showed that the increase in radiation intensity from Fuel 14 blended with indene ranged from 44 to 60 percent (averaged 52 percent) of the increase observed from Fuels 11 and 13 blended with methyl naphalene and tetralin.

FIGURE 25. EFFECT OF POLYCYCLIC AROMATIC RING CARBON ON FLAME RADIATION INTENSITY

VIII. DISCUSSION

It is the purpose of this discussion to (1) compare the observations made here with those observed in full-scale engine tests, and (2) form concepts to explain the effects of operating parameters on the sensitivities S_1 and S_3 to H/C ratio.

A. Comparison With Actual Engines

A favorable correlation between sooting tendency (smoke number, flame radia-
tion and liner temperature) and H/C ratio has been found for almost all gas
turbine engines. Figure 1 showed that the sooting tendency is strongly
dependent on engine design, but the sensitivities to H/C ratio are roughly
similar except for the striking difference exhibited by the J85 engine.(11)
When the full power operating conditions for the J85 engine are compared
with the J79 engine, as shown below, it is apparent that there are signifi-
cant differences in the burner inlet temperatures and reference velocities.

	J79-10A	J85-21
P	197 psia (1358 kPa)	121 psia (834 kPa)
T	736°F (664 K)	574°F (574 K)
F/A	0.0207	0.0196
V_{ref}	94 ft/s (28.7 m/s)	125 ft/s (38.1 m/s)
ρ	7.14 kg/m^3	5.07 kg/m^3

Calculations based on Equation 1 show that the sensitivity (S_1) for the J85
engine would be 2.5 times as large as that for the J79 engine at the opera-
ting conditions for takeoff. Actually, the sensitivity based on smoke
number (SN), i.e., the slope of ln(SN) versus H/C ratio line, for the J85
engine is about four times as large as that for the J79 engine so other
geometric or design factors are also important.

The results shown in Figure 23 for the increased sensitivity (S_3-S_1) to H/C
ratio caused by polycyclic aromatic content of the fuels suggest that the
effects of polycyclics in the fuel will be negligible at operating condi-
tions of high F/A ratio and low reference velocity. The T-63 combustor
tests (13) showed little or no sensitivity to polycyclic aromatics in the
fuel. This may have been due to the high F/A ratio (0.02) and the low
reference velocity (19 m/sec) used for the full power operating condition.
In general, engines operating at full power where sooting tendency is criti-
cal should not be expected to be highly sensitive to the molecular structure
of the aromatics because high F/A ratios are used at this condition. This
conclusion is consistent with several full-scale engine tests (11) except
for the TF41 results.(13)

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B. Concentration Effects of Polycyclic Aromatics

The various studies on the effects of polycyclic aromatics on soot formation include several anomalous results which may be attributed to concentration effects. Jackson and Blazowski(2) found that in particular fuel groups with the same hydrogen content (13 percent), the fuels containing high concentrations of tetralin exhibited higher than average liner temperature in a T-56 combustor at the cruise condition, but fuels containing equal amounts of both monocyclic and dicyclic aromatics did not form soot any more readily than fuels prepared with monocyclic aromatics exclusively. Similar conflicting results were obtained in studies (4) with the Phillips research combustor in which two fuels of essentially the same hydrogen content (13 percent) were prepared; the weight percent monocyclic and dicyclic ring carbon contents of one fuel were 10.5 and 4.9 and the others were 7.7 and 7.8, respectively. Both of the fuels gave the same flame radiation and exhaust smoke. More recent work (12), as well as the results presented here, show that the high concentrations (30 percent) of tetralin and dicyclic aromatics significantly increase the sooting tendency. Possibly, the reason for the insensitivities to polycyclic aromatics in the earlier studies (2, 4) is that the changes in concentration were relatively small and the effects on sooting tendency were buried in data scatter. The results in Figure 25 show that the increase in radiation intensity due to polycyclic aromatics is directly proportional to the polycyclic aromatic ring carbon content. However, the change is very slight for fuels that contain 13.5 percent hydrogen. In fact, the radiation intensities from Fuels 2 and 3 were only slightly higher than Fuel 1 in most of the experiments; the differences in their polycyclic and monocyclic aromatic ring carbons were greater than in the case of the earlier studies.(2, 4) The results show that the concentration of polycyclic aromatics must be quite high (10 percent in most cases) if any effect is to be observed.

C. Molecular Structure

The sensitivities S_2 and S_3 for fuels containing tetralin and dicyclic aromatics, respectively, were found to be equal. This does not infer that

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tetralin has the same sooting tendency as, say naphthalene, because more
tetralin had to be added to the base fuel to achieve a given hydrogen con-
tent than was the case for fuel blended with methyl naphthalenes. The
polycyclic aromatic content of the fuel should be expressed in terms of
aromatic ring carbon if it is polycyclic aromatic nuclei that are important
in increasing the sooting tendency by providing initial growth sites for
soot. If it is assumed that both rings in tetralin are aromatic, the dicy-
clic aromatic ring carbon in Fuels 2, 6, and 13 would average about 35
percent greater than the dicyclic ring carbon content of Fuels 3, 7, and 11.
This suggests that about 74 percent of the tetralin forms a naphthalene
intermediate during the pyrolysis phase of the soot-forming process. In an
earlier study (12), it appeared that tetralin was about 60 percent as ef-
fective as naphthalene in forming soot. In view of the fact that Benjamin,
et al. (23) showed that naphthalene was a major product of the pyrolysis of
tetralin, it stands to reason that tetralin would be more effective in
forming soot than indene where this conversion is unfavorable. However,
the effect of indene is difficult to compare with that of tetralin because
it has a lower hydrogen content, and smaller amounts of it were required in
blending Fuel 14 as compared with the amount of tetralin used in Fuel 13.
Indan would have been a better choice to compare with tetralin, but its cost
and availability were prohibitive. The results showed that Fuel 14 con-
taining indene had a higher sooting tendency than Fuel 8 containing mono-
cyclic aromatics. It is concluded that soot tendency of indene is between
that of alkyl benzenes and dicyclic aromatics.

D. Mechanism and Theory

The burner inlet temperature and reference velocity in the combustor had
strong effects on the sensitivities, S_1, S_2, and S_3 to H/C ratio. The
fuel/air ratio was less important, and the burner inlet gas density had a
negligible effect on the sensitivities.

Temperature appears to be a very important parameter in the formation of
soot. Millikan (10) showed that soot formation in premixed flames decreases
as the flame temperature is increased. This was explained by increased

oxidation of soot precursors by OH radicals in the flame. The OH radical concentration in the flame increased more rapidly with temperature than the soot precursor concentration, i.e., oxidation reactions dominated fuel pyrolysis reactions. On the contrary, work with diffusion flames by Glassman and Yaccarino (9) showed that the sooting tendency increased as flame temperatures increased. This work concluded that pyrolysis reactions are dominant in diffusion flames. In a recent study with the Phillips research combustor(13), it was found that the flame opacity in the primary zone increased as the burner inlet temperature was increased. Flame temperature measurements by the Kurlbaum technique showed that soot particle temperature increased with the corresponding increase in burner inlet temperature. These observations also suggest that fuel pyrolysis is a dominant process in the formation of soot in diffusion flames. However, the role of hydrogen content in the sooting tendency seems to be related to the oxidation reactions and the destruction of soot precursors by OH radicals. Several workers (7, 10, 24, 25) have alluded to the possibility that sources of hydrogen such as water in the flame zone reduce soot formation by increased oxidation by OH radicals.

These observations may explain the relatively strong effects of burner inlet temperature and reference velocity on the sensitivity to hydrogen content. If the soot-forming mechanism is described in terms of characteristic times for pyrolysis and oxidation, the effects of temperature and reference velocity become evident. When the reference velocity is increased, a higher level of turbulence is formed, which increases the mixing rate and raises the flame speed. This reduces the time allotted for pyrolysis and increases the probability of oxidizing soot precursors. As the temperature is increased, the rate of pyrolysis increases markedly and the extent of soot formation in the pyrolysis zone becomes greater. If the major soot-forming reactions become essentially complete within the pyrolysis zone, the effect of OH radicals on the destruction of soot precursors is relatively unimportant because the virgin soot particles are already formed and are relatively resistant to oxidative attack. Thus, when the velocity is high and the temperature is low, the characteristic time for pyrolysis is relatively short and the reaction rates in the pyrolysis zone are relatively slow. The

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oxidation processes involving OH radicals become important because the soot-forming reactions are still relatively incomplete; this results in a high sensitivity to hydrogen content ($1/R$ $[dR/d(H/C)]$).

In laminar diffusion flames, there is ample time for pyrolysis reactions because the only transport process is molecular diffusion. When the characteristic time for pyrolysis is relatively long, the effect of H/C ratio on the sooting tendency should be negligible. Recent studies (9, 26) on laminar diffusion flames conclude that sooting tendency correlates better with molecular structure than H/C ratio. When the effects of H/C ratio are relatively unimportant, the role of molecular structure is dominant because it greatly affects the pyrolysis reactions leading to soot. The opposite is generally true in highly turbulent flames in combustors because there, the pyrolysis is rate limited and the H/C ratio is the principal correlating parameter. This fact places some question on the validity of using results from laminar flame studies to predict combustor results.

The effect of pressure on the sensitivity ($1/R$ $[dR/d(H/C)]$) has been investigated by Friswell.(3) The sensitivities used by Friswell were determined from exhaust smoke measurements. He found that the sensitivity to hydrogen content peaked at about 0.5 MPa, and then decreased monotonically to essentially zero at about 2 MPa. The results of the present study show only a small effect of gas density on the sensitivity to hydrogen content. It has been argued (12) that thermolecular reactions are significant in the formation of soot, and probably play their most important role in the pyrolysis phase of the mechanism. If these reactions increase the rate of pyrolysis relative to that of oxidation, it follows that high densities or pressures would reduce the sensitivity to hydrogen content.

The work presented here shows that changes in gas density have very little effect on the sensitivity of the sooting tendency to hydrogen content. However, the very subtle differences in the dependencies of S_1 and S_3 indicate an effect of density on the sensitivity to polycyclic aromatics. Earlier work (12) showed a similar effect, i.e., sensitivity to polycyclic aromatics decreased with increasing pressure, but the peak in the sensi-

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tivity shown in Figure 23 was not evident. Similarly, while the effect of fuel/air ratio on S_1 and S_3 is modest, the effect on the sensitivity to polycyclic aromatics (S_3-S_1) is very apparent.

The effects of operating parameters on the sensitivities to H/C ratio appear to be explainable in terms of the global relative rates and characteristic times for pyrolysis and oxidation reactions. In accordance with other studies (1-5, 12), the results show that H/C ratio is the principal correlating parameter for the sooting tendency, while the effect of polycyclic aromatics is seen as a perturbation of this correlation. The increased sooting tendency of polycyclic aromatics appears to be related to differences in the pyrolysis reactions. It has been proposed that the formation of soot depends on the formation of polycyclic aromatic molecules which serve as growth sites or soot precurors (12). Recently (27), single-, double-, and triple-ring aromatic structures have been observed as precursors to soot formation in rich methane/oxygen flames. Obviously, then, aromatic structures are formed rapidly and abundantly in the pyrolysis of even the most simple fuel molecules. When polycyclic aromatics are initially present in the fuel, a significant number of these structures appear to survive the fragmentation reaction channels that form lower molecular weight intermediates. Monocyclic aromatics follow a similar route, but to a lesser extent. The increased population of growth sites increases the probability of forming soot. However, the fragmentation products are also necessary in forming key soot precursors such as diacetylene, butadiene, and other possible C_4 structures which may form cyclic adducts on existing aromatic growth sites. The bulk of the soot formed may well be composed of fragmentation products because these species would appear to be destroyed by oxidation reactions more easily than growth sites which have a relatively high molecular weight and a compact structure. Without the low molecular weight species, the sensitivity to the H/C ratio may be much smaller.

The subtle differences in the effects of operating conditions on the sensitivity S_3-S_1 to polycyclic aromatics (see Figure 24) is basically caused by these differences in rate processes which are dependent on fuel composition.

IX. CONCLUSIONS

The results from the Phillips 2-inch combustor indicate that:

- In gas turbine flames, there is relatively little time for fuel pyrolysis before soot precursors become subject to oxidation. The H/C ratio of the fuel appears to be important because of its effect on the OH radical concentration.

- In gas turbine combustion, the operating conditions are important in determining fuel sensitivity because they affect the amount of fuel pyrolysis which occurs before oxidative attack begins. Low reference velocities and high temperatures tend to favor conversion of soot precursor to soot nuclei to more stable soot particles, which are more resistant to oxidation. This reduces the sensitivity to H/C ratio.

- Under some operating conditions, the sooting tendency is significantly affected by polycyclic aromatics including tetralin, but the H/C ratio is still the principal correlating parameter. At high power conditions where soot formation is of greatest concern, the fuel/air ratio of most engines is high (0.02), and there is little sensitivity to polycyclic aromatics.

- In the Phillips combustor, the relatively small changes in flame temperature caused by variations in fuel composition (H/C ratio) do not account for the significant effect that hydrogen content has on the sooting tendency.

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LIST OF ABBREVIATIONS AND SYMBOLS

F/A	=	Fuel/air weight ratio
T	=	Burner inlet temperature, K
V	=	Reference velocity, m/sec
ρ	=	Gas density, kg/m^3
R	=	Flame radiation intensity, kW/m^2
H/C	=	Hydrogen/carbon atom ratio
S	=	Sensitivity of sooting tendency to H/C ratio, $1/R \left[dR/d(H/C) \right]$
SN	=	Smoke number
ϵ	=	Combustion efficiency
UBH	=	Unburned hydrocarbons
LMN	=	Liquid methyl naphthalenes
BTX	=	Benzene, Toluene, Xylene
CC	=	Correlation Coefficient

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